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## (54) INTERLAYER FILM FOR LAMINATED GLASS AND LAMINATED GLASS

## (57)Abstract:

PROBLEM TO BE SOLVED: To provide an interlayer film for laminated glass which has excellent mechanical property, transparency, in particular, haze, thermal insulation and electromagnetic wave transmission and shows excellent resistance to penetration in the lapse of time when being fabricated into a laminated glass and to provide a laminated glass.

SOLUTION: In this interlayer film for laminated glass, a polyvinyl acetal resin, phyllosilicate, plasticizer, adhesive power adjusting agent, ITO fine particle and dispersant are included and the ITO fine particle and laminar silicate are dispersed microscopically-uniformly. The laminated glass is also provided.

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## CLAIMS

- [Claim(s)]
- [Claim 1] An interlayer, wherein it contains polyvinyl-acetal resin, a sheet silicate, a plasticizer, an adhesive strength regulator, ITO (tin dope indium oxide) particles, and a dispersing agent and uniform dispersion of ITO particles and the sheet silicate is carried out minutely.
- [Claim 2] Polyvinyl-acetal resin 100 weight section, 0.05 to sheet silicate 20 weight section, At least one or more kinds of 0.0001 to metal salt 1.0 weight sections chosen from a group which consists of 20 to plasticizer 100 weight section, alkali metal salt, and alkaline earth metal salt, The interlayer for glass laminates according to claim 1 consisting of 0.1 to ITO particle 3.0 weight section, and 0.001 to dispersing agent 5.0 weight section, distributing and becoming further so that sheet silicates with a size of 1 micrometers or more may be ten or less 100 micrometer<sup>2</sup> hits.
- [Claim 3] Mean particle diameter is 80 nm or less, and ITO particles in a film are the particle numbers of not less than 100 nm. The interlayer for glass laminates according to claim 1 or 2 currently distributing so that it may become one piece less than 1 micrometer<sup>2</sup>.
- [Claim 4] The interlayer for glass laminates according to any one of claims 1 to 3, wherein a sheet silicate is an organically-sized sheet silicate.
- [Claim 5] The interlayer for glass laminates according to any one of claims 1 to 4, wherein a dispersing agent is at least one or more sorts chosen from a group which consists of a phosphoric ester system, recinoleic acid, and polyrecinoleic acid.
- [Claim 6] The interlayer for glass laminates according to any one of claims 1 to 5, wherein polyvinyl-acetal resin is polyvinyl butyral resin.
- [Claim 7] A glass laminate using the interlayer for glass laminates according to any one of claims 1 to 6.
- [Claim 8] The glass laminate according to claim 7 with which Hayes is characterized by visible light transmittance's being not less than 70%, and solar transmittance (300 nm ~ 2500 nm) being 80% or less of visible light transmittance 1.0% or less.

[Translation done.]

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## DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the glass laminate which uses the interlayer for glass laminates and it which were excellent in transparency, thermal insulation nature, and electromagnetic wave permeability, and adhesive strength with glass is suitable for, and were excellent also in penetration resistance.

[0002]

[Description of the Prior Art] Conventionally, since it is rare for the fragment of glass to disperse and it is safe even if it damages in response to an external shock, the glass laminate is widely used as windowpanes, such as a vehicle like a car, an airplane, and a building, etc. Generally what makes the interlayer for glass laminates which consists of polyvinyl-acetal resin, such as polyvinyl butyral resin plasticized with the plasticizer, intervene between the glass of a couple at least as the above-mentioned glass laminate, makes unity, and is obtained is used. The art of the interlayer in which the sheet silicate was minutely distributed in the film is also indicated by JP.2001-58853.A as an interlayer excellent in intensity, pliability, transparency, etc. Although the requests to the glass laminate which was excellent in the thermal insulation nature to which importance was seldom attached until now as a glass laminate for a car or buildings on the other hand were mounting, although the above conventional glass laminates were excellent in safety, they had the problem of being inferior to thermal insulation nature.

[0003] Generally, although an amount of energy is as small as about 10% compared with ultraviolet rays, since a thermal effect is large, and is absorbed by the substance and the infrared rays which have the wavelength of not less than 780 nm also in a beam of light bring about a rise in heat, they are called the heat ray. Therefore, in order to improve thermal insulation nature, the method of intercepting the windshield of a car, side glasses, and the infrared rays containing the glass window and glass door of a building is examined, and by for example, vacuum evaporation, sputtering processing, etc. The heat ray cut glass etc, which provided coating layers, such as metal or a metallic oxide, in the glass surface, and gave thermal insulation nature are marketed. This coating layer was weak from the outside to the abrasion, and since chemical resistance was also inferior, the method of laminating interlayers, such as a plasticization polyvinyl-butylal resin film, and using as a glass laminate, for example was adopted.

[0004] However, the heat ray cut glass in which interlayers, such as the above-mentioned plasticization polyvinyl-butylal resin film, were laminated. There was a problem that transparency (visible light transmittance) falls since it is expensive and the multilayer coating tip is thick, or the adhesive property of a multilayer coating tip and an interlayer fell, and exfoliation and the white bluish mark of an interlayer happened. They are various kinds of communication equipment (3.5 MHz), for example, a ham radio, in recent years. Although urgency traffic apparatus, (10 MHz or less), VICS (a car information communications system, 2.5 GHz), ETC (a toll road electronic toll collection system, 5.8 GHz), satellite broadcasting (12 GHz), etc. are increasingly carried in a car, [ 7 MHz, ] The above-mentioned multilayer coating tip layer checked the penetration of electromagnetic waves, and had problems, such as interfering with communication functions, such as a cellular phone, car navigation, a garage opener, and a fee automatic acceptance system. A heat barrier is not provided in a glass surface, but the glass laminate which laminated the polyester film which vapor-deposited metal between the interlayers for glass laminates is indicated by JP.61-52093.B, JP.64-36442.A, public law,

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etc. However, there were problems, like exfoliation not only takes place, but as for a glass laminate of the above-mentioned indication, there is a problem in the adhesive property between a plasticization polyvinyl-butylal-resin sheet and polyester film, and the interface of an electromagnetic wave penetration is also insufficient, and it is by it.

[0005]

[Problem(s) to be Solved by the Invention] Intensity and pliability are compatible, and transparency, especially Hayes are good, and the purpose of this invention is to provide the glass laminate using the interlayer for glass laminates excellent in thermal insulation nature, and this interlayer for glass laminates, without moreover reducing electromagnetic wave permeability.

[0006]

[Means for Solving the Problem] This invention contains polyvinyl-acetal resin, a sheet silicate, a plasticizer, an adhesive strength regulator, ITO (tin dope indium oxide) particles, and a dispersing agent, and ITO particles and a sheet silicate are the interlayers by which uniform dispersion is carried out minutely.

[0007] In this invention, by using ITO particles, in order to raise thermal insulation nature, and carrying out uniform dispersion of this ITO particle minutely in a film, transparency (especially Hayes) and electromagnetic wave permeability were able to be reduced, and thermal insulation nature was able to be raised without things. Intensity and pliability were able to be reconciled in this invention, without reducing transparency (especially Hayes), since a sheet silicate is used and uniform dispersion is minutely carried out into a film.

[0008] If polyvinyl-acetal resin used by this invention is polyvinyl-acetal resin produced by acetalizing polyvinyl alcohol (PVA) resin by aldehyde, it will not be limited in particular. The above-mentioned PVA resin is obtained by usually saponifying polyvinyl acetate, and, as for a saponification degree, 80-99.8-mol% of PVA resin is generally used. Although in particular a molecular weight and molecular weight distribution of polyacetal resin that are used for this invention are not restricted, from a moldability, physical properties, etc., a thing of 200-3000 is preferably used for a degree of polymerization of PVA resin used as a raw material, and resin of the degrees of polymerization 500-2000 is used especially preferably. If the penetration resistance of a glass laminate obtained as the above-mentioned average degree of polymerization is less than 200 falls and the above-mentioned average degree of polymerization exceeds 3000, the moldability of a resin layer worsens, moreover, the rigidity of a resin layer will become large too much, and processability will worsen.

[0009] As long as are used, and a carbon number chooses suitably, and is used according to performance demanded, its \*\*\*\* is good and aldehyde of 1-10 is required for it as the above-mentioned aldehyde, two or more kinds may be used together. As an example of aldehyde, for example n-butylaldehyde, isobutylaldehyde, N-valeraldehyde, 2-ethylbutylaldehyde, n-hexylaldehyde, n-octyl aldehyde, n-nonyl aldehyde, n-decyl aldehyde, formaldehyde, acetaldehyde, benzaldehyde, etc. are mentioned. As aldehyde used preferably, n-butylaldehyde, n-ethylaldehyde, and n-valeraldehyde are mentioned. A carbon number is butylaldehyde of 4 especially preferably.

[0010] As desirable polyvinyl-acetal resin, polyvinyl-butylal (PVB) resin acetalized with butylaldehyde is mentioned especially. After these acetal resin takes required physical properties into consideration, it may be blended in a suitable combination. It is also possible \*\* polyvinyl-acetal resin which combined two or more kinds of aldehyde at the time of acetalization, and to use suitably. The degree of acetalization of said polyvinyl-acetal resin used by this invention is usually 40 to 85%, and is 60 to 75% preferably.

[0011] A sheet silicate used by this invention has an exchangeable cation between layers of a detailed flaky crystal about 1 nm thick. In [ are the silicate mineral condensed in layers by an ionic bond, and ] this invention, The layer structure is exfoliated by a chemical or physical means, by distributing this flake uniformly in a transparent resin composition, the transparency of a resin composition would be held and also a function as an inorganic bulking agent, a filler, and a viscosity controlling agent can be exhibited in a resin composition.

[0012] Although a kind in particular of the above-mentioned sheet silicate is not limited,

montmorillonite, There is a vermiculite, halloysite, or swelling mica besides smectite system argillite, such as saponite, hectorite, beidelite, a stevensite, and nontronite, etc., and what was compounded [ a natural thing or ] can be used preferably.

[0013] As shape of a sheet silicate used for this invention, 0.01-3 micrometers and thickness are

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used for average length, a thing of 20-500 is preferably used for 0.001-1 micrometer and an aspect ratio is more suitably, 0.05-2 micrometers and thickness are used for average length, and a thing of 50-200 is used for 0.01-0.5 micrometer and an aspect ratio.

[0014] Although cation exchange capacity in particular of a sheet silicate used for this invention is not limited, it is preferred that it is 50 to 200 mmol/100g. In a case below 50 mmol(s)/100g, quantity of a plasticizer and a cation system surface-active agent which are intercalated by ionic exchange between crystal layers decreases easily, and a sheet silicate may not be distributed minutely as a result. On the other hand, it may be difficult for associative strength between layers of a sheet silicate to become firm, and for INTAKARE by plasticizer and a cation system surface-active agent to become insufficient, and to distribute a sheet silicate minutely, in exceeding 200 mmol/100 g.

[0015] Although a sheet silicate may be used as it is and an organically-ized sheet silicate organically-ization-processed beforehand may be used, it is preferred to use an organically-ized sheet silicate. Under the present circumstances, even if a sheet silicate which is not organically-ized exists in an organically-ized sheet silicate, it is satisfactory in any way. Since between layers of a sheet silicate is easy to be distributed more minutely in resin than a sheet silicate as for which organically-ization is a sheet silicate which it comes to process and which is not organically-ized with a cation system surface-active agent, the above-mentioned organically-ized sheet silicate is used more suitably. As a cation system surface-active agent used for organically-ization, quarternary ammonium salt, the 4th class phosphonium salt, etc. are mentioned, and quarternary ammonium salt which has at least one with a carbon numbers of eight or more alkyl chain preferably is used. When it does not contain an alkyl chain whose carbon number is eight or more, the hydrophilic nature of alkyl ammonium ion cannot be strong, and cannot fully un-polarize between layers of a sheet silicate. As quarternary ammonium salt which has a with a carbon numbers of eight or more alkyl chain, For example, lauryl trimethylammonium salt, a stearyl trimethyl AMMONIMU salt, triocetyl ammonium salt, a distearyl dimethylbenzylammonium salt, a JI hardening beef tallow dimethylammonium salt, distearyl dibenzylammonium salt, etc. are mentioned.

[0016] As for an addition of a sheet silicate, it is preferred that they are 0.05 to 20 weight sections to polyvinyl-acetal resin 100 weight section. In less than 0.05 weight sections, when it is few, and an addition does not fully come to demonstrate desired physical properties but adds exceeding 20 weight sections, a pitch which occupies in an increase composite material may decrease, a fall of physical properties, such as a fall of transparency, aggravation of Hayes, and shock resistance, etc. may be produced, and it is not desirable. An addition of a more desirable sheet silicate is 0.5 to 5 weight section.

[0017] A sheet silicate needs to distribute minutely and it is not preferred on mechanical strength, especially transparency that many sheet silicates with a viewing and a size of 1 micrometers or more which can be checked on a scanning electron microscope (SEM) level exist as a grade of the distribution. Quantity of a sheet silicate of 1 micrometers or more or an organically-ized sheet silicate is ten or less per 10 micrometers x 10 micrometers, and a desirable dispersion state is five or less pieces still more preferably.

[0018] Especially as a method of distributing a sheet silicate minutely in a film, although not limited, especially a thing for which a sheet silicate and a plasticizer are mixed beforehand, and what fully swelled an interlayer spacing of a sheet silicate is added and kneaded to resin is preferred. By mixing a sheet silicate and a plasticizer beforehand, it is because the time of a plasticizer swelling and said sheet silicate being mixed with resin becomes easy to be distributed in resin minutely easily. In this case, some plasticizers and the whole quantity of a sheet silicate are once mixed, and a residue of a plasticizer may be applied further after that and it may mix.

[0019] As a plasticizer, especially if conventionally used for an interlayer or polyvinyl-acetal resin, it is not limited, and phosphoric acid system plasticizers, such as organic acid ester system plasticizers, such as monobasic-organic-acid ester and polybasic organic acid ester, an organic phosphorus acid system, and an organic phosphorous acid system, etc. are used, for example. As a monobasic-organic-acid ester system plasticizer, for example Triethylene glycol, Glycol and butanoic acid, such as tetrachethylene glycol and tripropylene glycol. Isobutyric acid, caproic acid, 2-ethylbutanoic acid, heptylacid, n-octylic acid, Glycol system ester obtained by a reaction with monobasic organic acid, such as 2-ethylhexyl acid, pelargonic acid (n-nonylic acid), and decylacid, is mentioned. Triethylene glycol -\*\*\*\* prong acid ester, triethylene glycol di-2-ethylbutylate especially, Monobasic-organic-

acid ester of triethylene glycols, such as triethylene glycol di-n-octyl acid ester and triethylene glycol di-2-ethylhexyl acid ester, is used suitably. As a polybasic organic acid ester system plasticizer, For example, straight chain shape of polybasic organic acid, such as adipic acid, sebacic acid, and azelaic acid, and the carbon numbers 4-8 or ester with letter alcohol of branching is mentioned. Especially, dibutyl sebacic acid ester, dioctylazelaic, dibutylcarbitol adipate, etc. are used suitably. As an organic phosphorus acid system plasticizer, tributoxyethyl phosphate, isodecyl phenyl phosphate, triisopropyl phosphate, etc. are mentioned, for example. One kind is used independently, \*\*\*\* of the above-mentioned plasticizer is also good, and two or more kinds may be used together.

[0020] As an example of a plasticizer used especially preferably, For example, triethylene glycol -\*\*\*\* prong acid ester, triethylene glycol di-2-ethylbutylate, triethylene glycol di-n-octyl acid ester, triethylene glycol di-2-ethylhexyl acid ester, etc. are mentioned. These plasticizers are properly used in consideration of compatibility with resin, etc. according to a kind of polyvinyl-acetal resin.

[0021] An addition of a plasticizer has 20 to 100 preferred weight section to polyvinyl-acetal resin 100 weight section. Less than 20 weight sections are insufficient for distributing a sheet silicate minutely, and penetration resistance may fall. When a plasticizer is added exceeding 100 weight sections, bleed out of a plasticizer arises and there is a possibility that optical strain of a glass laminate obtained by the transparency of a resin layer and an adhesive property falling may become large. An addition of a desirable plasticizer is 30 to 60 weight section.

[0022] At least one or more kinds of metal salt chosen from a group of this invention which makes it meet and becomes an interlayer for glass from alkali metal salt and alkaline earth metal salt as an adhesive strength regulator is used. Especially as the above-mentioned metal, it is not limited, for example, sodium, potassium, magnesium, etc. are mentioned. As acid which constitutes the above-mentioned salt, inorganic acid, such as organic acid, such as octylic acid, hexylacid, butanoic acid, acetic acid, and formic acid, or chloride, and nitric acid, is mentioned, and a salt of organic acid of the carbon numbers 2-16 is used suitably especially. As still more desirable metal salt, they are carboxylic acid magnesium salt of the carbon numbers 2-16, or carboxylic acid potassium salt of the carbon numbers 2-16. It is not limited especially as carboxylic acid magnesium salt of the carbon numbers 2-12, or carboxylic acid potassium salt of the carbon numbers 2-12. For example, magnesium acetate, potassium acetate, magnesium propionate, Potassium propionate and magnesium 2-ethylbutanoate and potassium 2-ethylbutanoate, 2-hexanoic acid magnesium, 2-ethylhexanoic acid potassium, etc. may be used preferably, and these may be used independently, or two or more sorts may be used together.

[0023] [0.0001 to 1.0 weight section is desirable still more preferred to polyvinyl-acetal resin 100 weight section, and an addition of at least one or more kinds of metal salt chosen from a group which consists of the above-mentioned alkali metal salt and alkaline earth metal salt is 0.01 to 0.2 weight section. In less than 0.0001 weight sections, an adhesive strength fall of a periphery takes place under a high humidity atmosphere, and if 1.0 weight sections are exceeded, adhesive strength will become low too much, and also a problem that membranous transparency falls will arise.

[0024] In this invention, in order to give thermal insulation nature, ITO particles are used. It is not limited that what is necessary is [especially] just the ITO particles which dope tin to indium oxide and are usually used for addition of conductivity as ITO particles. As particle diameter of ITO particles, 100 nm or less has preferred mean particle diameter of a primary particle. When it exceeds 100 nm, transparency may fall.

[0025] Content of ITO particles has 0.1 to 3.0 preferred weight section to polyvinyl-acetal resin 100 weight section. Content becomes difficult to come out of an infrared cutting effect in less than 0.1 weight sections, thermal insulation nature will fall, if 3.0 weight sections are exceeded conversely, the permeability of visible light will fall, and Hayes will also become large.

[0026] It is necessary to carry out uniform dispersion of the ITO particles minutely into an interlayer, and when an ITO particle kind is not distributing uniformly minutely, transparency (especially Hayes) will fall. It is preferred to distribute that mean particle diameter is 80 nm or less, and a particle number of not less than 100 nm is below one piece/1micrometer<sup>2</sup> as a dispersion state in inside of an interlayer of ITO particles.

[0027] It is not limited especially as a method of distributing ITO particles in resin, but after making it usually distribute uniformly in carrier fluid which consists of organic solvents, it is made to distribute in resin and uses, but in this invention, it is preferred to make it distribute as carrier fluid using a

plasticizer used for an interlayer and a plasticizer of the same kind.

[0028]In this invention, in order to carry out uniform dispersion of the ITO particles minutely into resin, a dispersing agent is used. Uniform dispersion of the ITO particles can be minutely carried out into a film, and membranous Hayes can be made to improve further by using a dispersing agent. It may be beforehand added by carrier fluid and a dispersing agent may be used for it, when mixing ITO particles with resin, it may be added, and it may be used.

[0029]As a dispersing agent, dispersing agents generally used as a dispersing agent of inorganic particles, such as a compound which has a sulfate ester system compound, a phosphoric ester system compound, carboxylate, a polyhydric alcohol type surface-active agent, and at least one or more carboxyl groups, are mentioned. A phosphoric ester system compound and a compound which has at least one or more carboxyl groups are suitably used as a dispersing agent among these. As a phosphoric ester system compound, alkyl phosphate, a polyoxyethylene alkylphenol ether phosphate, polyoxyethylene-alkyl-ether phosphoric acid, etc. are mentioned, for example. As a compound which has at least one or more carboxyl groups, hydroxy acid especially recinoic acid, and polyrecinoic acid are used suitably.

[0030]0.001 to 5.0 weight section is desirable still more preferred to polyvinyl-acetal resin 100 weight section, and quantity of the above-mentioned dispersing agent is 0.005 to 3.0 weight section. When quantity of a dispersing agent is less than 0.001 weight sections, and the addition effect can hardly expect but exceeds 5.0 weight sections, there is a possibility that it may be alike at the time of film production and glass laminate creation, and may foam, or adhesive strength of an interlayer and glass may be improved too much.

[0031]When distributing ITO particles, a distributed auxiliary agent may be used in addition to a dispersing agent, and a compound which has at least one or more carboxyl groups except the above-mentioned recinoic acid and polyrecinoic acid as a distributed auxiliary agent, a chelating agent, etc. are mentioned. Although it is possible not to limit and to use EDTA and beta-diketones especially as the above-mentioned chelating agent, What has good compatibility with a plasticizer or resin is preferred, beta diketones, such as an acetylacetone, benzoyltrifluoroacetone, and dipivaloyl methane, are desirable still more preferred also especially in a chelating agent, and an acetylacetone is used suitably. When these chelating agents configure to ITO particles, it is thought by barring condensation of ITO particles that Hayes improves. As an addition of the above-mentioned chelating agent, 0.001 - the amount part of duplexes are preferred to polyvinyl-acetal resin 100 weight section, and it is 0.01 to 1 weight section more preferably. When the amount part of duplex is exceeded, there is a possibility of foaming at the time of film production, or producing foaming at the time of glass laminate production. An effect can hardly be expected to be less than 0.001 copy.

[0032]As a compound with one or more carboxyl groups except the above-mentioned recinoic acid and polyrecinoic acid, Aliphatic carboxylic acid, aliphatic dicarboxylic acid, aromatic carboxylic acid, aromatic dicarboxylic acid, hydroxy acid, etc. are mentioned, and benzoic acid, phthalic acid, salicylic acid, etc. can specifically be used. Especially, aliphatic carboxylic acid of C2-C18 and hydroxy acid are used suitably, and are aliphatic carboxylic acid of C2-C10 more preferably. Specifically, acetic acid, propionic acid, n-butanolic acid, 2-ethylbutanoic acid, n-hexanoic acid, 2-ethylhexanoic acid, n-octanoic acid, etc. are mentioned.

[0033]As a quantity of a compound with one or more carboxyl groups except the above-mentioned recinoic acid and polyrecinoic acid, it is 0.001 - the amount part of duplex to polyvinyl-acetal resin 100 weight section, and a more desirable addition is 0.01 to 1 weight section. When the amount part of duplex is exceeded, there are fear of membranous yellowing and a possibility of spoiling adhesive strength of glass and a film, and the addition effect is hard to accept to be less than 0.001 weight section.

[0034]Additive agents, such as an antioxidant, an ultraviolet ray absorbent, lubricant, fire retardant, a spray for preventing static electricity, an adhesive strength regulator, \*\*\*\*\* a heat ray reflective agent, and a heat ray absorbent, may be added by interlayer for glass laminates of this invention if needed in the range which does not check an effect of the invention.

[0035]Although thickness in particular of an interlayer for glass laminates of this invention is not what is limited -- as a glass laminate -- the minimum -- when required penetration resistance and weatherability are taken into consideration, it is preferred that it is 0.3-0.8 mm practical. Improvement in penetration resistance, etc. may laminate and use the interlayers for glass laminates

of this invention or an interlayer for glass laminates of this invention, and other interlayers if needed. [0036]Although obtained by an interlayer of this invention laminating the above-mentioned interlayer for glass laminates, and glass, especially as glass, it is not limited but transparent plate glass currently generally used can be used. Although usual glass may be used as glass, heat absorbing glass which intercepts infrared rays of a wavelength area (900 nm - 1300 nm) is preferred, and especially heat absorbing glass whose transmissivity in a full wave length field (900 nm - 1300 nm) is 65% or less is preferred. Since infrared cutoff performance of ITO particles is large at the long wavelength side and comparatively smaller than 1300 nm in a wavelength area (900 nm - 1300 nm). By combining with glass which absorbs infrared rays of a wavelength band (900 nm - 1300 nm), wide range infrared rays can be intercepted and the thermal insulation effect can be heightened. As glass used as a glass laminate, what is called organic glass, such as polycarbonate, polymethylmethacrylate, etc. which were excellent in transparency in addition to inorganic glass, may be used.

[0037]What is necessary is not to be limited especially as a method of obtaining an interlayer for glass laminates of this invention, to knead polyvinyl-acetal resin, a sheet silicate, a plasticizer, an adhesive strength modifier, ITO particles, and a dispersing agent, and just to produce a film. Under the present circumstances, especially thing of a sheet silicate for which what was beforehand distributed in a plasticizer is added and kneaded to resin is preferred. By mixing a sheet silicate and a plasticizer beforehand, when a plasticizer swells in said sheet silicate, between layers of a sheet silicate spreads and it mixes with resin, easily, into resin, it distributes minutely and burns -- it is because it becomes. In this case, some plasticizers and the whole quantity of a sheet silicate are once mixed, and a residue of a plasticizer may be applied further after that and it may mix. As point \*\* also of the ITO particles was carried out, adding, after carrier fluid has distributed beforehand is preferred.

[0038]Although a device in particular that mixes a plasticizer, a sheet silicate and ITO particles, a plasticizer, and a dispersing agent is not limited, planet type agitating equipment, a wet mechanochemical device, a Henschel mixer, a homogenizer, an ultrasonic irradiation machine, etc. are generally used. Although a device used for kneading of polyvinyl-acetal resin, a sheet silicate, a plasticizer, an adhesive strength modifier, ITO particles, and a dispersing agent is not limited, either, an extrusion machine, plastograph, a kneader, a Banbury mixer, a calendaring roll, etc. can be used. It is preferred to use an extrusion machine from a viewpoint of producing continuously especially.

Although it is not limited especially as a method of fabricating an interlayer for glass laminates of this invention but what is necessary is just to produce a film by extrusion process, the calendar method, the pressing method, etc. it can be more preferably based on an extrusion process by the biaxial said direction, and Hayes can be made to improve further.

[0039]As for a glass laminate of this invention, it is preferred that 1.0% or less and visible light transmittance are not less than 70%, and Hayes is [solar transmittance] 80% or less of visible light transmittance. Visible light refers to light whose wavelength is 380-780 nm, and the solar radiation transmitted light is a light whose wavelength is 300-2500 nm here. If visible light transmittance is not less than 70%, it comes out and there is Hayes of a glass laminate at 1.0% or less, it exceeds in transparency, and if solar transmittance (300-2500 nm) is 80% or less of visible light transmittance, it has the thermal insulation nature which was excellent since transmissivity of light of an infrared area by the side of long wavelength decreased rather than visible light.

[0040]A glass laminate using an interlayer for glass laminates of this invention can be used conveniently for a glass part of vehicles, such as a windshield of a car and side glasses, an airplane, and a train, structural glass, etc. It laminates with other films and it is also possible by using to use for example, as functional glass laminates, such as an insulation glass laminate which gave insulation etc. An interlayer for glass laminates of this invention is laminated with rigid bodies other than glass, for example, metal, an inorganic material, etc., and application as a damping raw material is also possible for it.

[0041](OPERATION) When equivalent to a visible light wavelength or high visible light transmittance usually adds an additive agent of size beyond it to resin of transparency like an indispensable interlayer like a sheet silicate of bulk, with an additive agent, visible light is scattered about strongly and decline in visible light transmittance, aggravation of Hayes, etc. pose a problem. However, since it is possible in this invention to distribute a sheet silicate minutely efficiently when a plasticizer invades between layers when distributing a sheet silicate in resin as mentioned above, Since

transparency is secured and it is not necessary to remove a plasticizer, an interlayer for glass laminates of outstanding physical properties is obtained easily. That is, an interlayer for glass laminates which is the purpose of original inorganic substance addition and which refining (coexistence of mechanical-strength intensity and pliability) of an interlayer of became possible, and was excellent in transparency, and reconciled mechanical-strength intensity and pliability is obtained, with high visible light transmittance maintained.

[0042] On the other hand, it is transparent, and an interlayer with the thermal insulation effect is obtained, holding transparency, since ITO particles which moreover have infrared absorption ability were minutely distributed by interlayer. Since ITO particles are smaller than visible light enough since it distributes uniformly in the state of a nano-scale ultrathin particle, and dispersion is not caused, an interlayer excellent in transparency, especially a haze value is obtained. Since ITO particles are minutely distributed in an interlayer with a gestalt of an ultrathin particle, it differs from \*\*\* glass and a heat ray reflective glass laminate using heat ray reflective PET by vacuum evaporation or coating which were conventionally used for a thermal insulation nature interlayer. When reflection of a communication wavelength belt does not take place but it is considered as a glass laminate, to communication functions, such as a cellular phone, car navigation, and a garage opener, it does not become a problem at all. By distributing a plasticizer beforehand and furthermore, using an ITO ultrathin particle, it can process like a manufacturing method of the usual interlayer, processability, workability, productivity, etc. are not spoiled, and it is obtained as usual.

[0043] The more a sheet silicate generally distributes a sheet silicate minutely in resin as a secondary effect with addition sweet red bean soup with mochi, the more a mechanical strength of a thermoplastics-sheet silicate composite, gas barrier property, and transparency improve remarkably. When an interface product of a sheet silicate and resin increases with improvement in distribution of a sheet silicate can explain. That is, polymer intensity can be efficiently increased, so that a distributed degree of a sheet silicate improves, since dynamics intensity, such as an elastic modulus of polymer, increases by restraining molecular motion of polymer in an interface of resin and an inorganic crystal. Since a resin layer tends to diffuse a gas molecule far as compared with an inorganic substance, when a gas molecule diffuses inside of a composite material, it is spread bypassing an inorganic substance. Therefore, a gas barrier can be efficiently raised, so that a distributed degree of a sheet silicate improves. By composite-izing an ITO ultrafine particle and a sheet silicate with a nano-scale as mentioned above, it becomes possible to give various functions to an interlayer.

[0044]

[Example] Hereafter, the contents of this invention are explained based on an example and a comparative example.

Examples 1-9, the comparative examples 1-4, 6-7 [Composition of a polyvinyl butyral] The heating and dissolving of 275 g of the PVA resin (average-degree-of-polymerization 1700 and saponification degree % of 99.2 mol) were added and carried out to the pure water 2890g. Temperature control of the system of reaction was carried out to 15 \*\*, 35% of the weight of the chloride 201g and the *n*-butylaldehyde 157g were added, this temperature was held, and the reactant was deposited. Then, hold the system of reaction at 80 \*\* for 3 hours, and the reaction was made to complete, superfluous water washed, unreacted *n*-butylaldehyde was flushed, sodium hydroxide solution neutralized the chloride catalyst, and white powdered PVB resin was obtained through rinsing and desiccation for 2 hours with still more superfluous water. The degree of formation of average butyral of this resin was 68.5-mol %.

[0045][Production of an ITO distribution plasticizer] As opposed to triethylene glycol di-2-ethyl HEKISHI rate 20 weight section, ITO powder (mean particle diameter of primary particle; 30 nm) 1 weight section was taught, and ITO particles were distributed in the plasticizer in the level type micro bead mill, using phosphoric ester of nonylphenyl polyethylene oxide as a dispersing agent. Then, it added under stirring of acetylacetone 0.1 weight section in the solution concerned, and the ITO distribution plasticizer was produced. The mean particle diameter of the ITO particles of a distributed plasticizer was 35 nm.

[Production of a sheet silicate distribution plasticizer] Triethylene glycol di-2-ethyl HEKISHI rate 20 weight section of a sheet mica (trade name MAE CO-OP CHEMICAL CO., LTD. make, organicity-processed processing article) 1 weight section were mixed for 1 minute with planet type agitating

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equipment, and the paste state silicone distribution plasticizer was obtained.

[0046][Manufacture of the interlayer for glass laminates] PVB resin 100 weight section obtained above, the specified quantity of an ITO distribution plasticizer in which ITO particles become the quantity of Table 2. The specified quantity of a sheet silicate distribution plasticizer in which a sheet silicate becomes the quantity of Table 2. The specified quantity of the plasticizer that the total amount of a plasticizer (triethylene glycol di-2-ethyl HEKISHI rate) becomes 40 weight sections, After fully carrying out melt kneading of the specified quantity of 2-ethylbutanoic acid magnesium in which magnesium content is furthermore set to 60 ppm to the whole system with a roll mill, press forming was carried out for 30 minutes at 150 °C using the press-forming machine, and the interlayer of 0.76 mm of average thickness was obtained. The mean particle diameter of the ITO particles in a film is 56 nm, and, as for a not less than 100-nm particle, particle diameter was not observed. A sheet silicate of 1 micrometers or more was not observed.

[0047] [Manufacture of a glass laminate] The interlayer for glass laminates obtained above is put from the both ends with a transparent float glass (30 cm by 30 cm×2.5 mm in thickness). After putting this in the rubber back and carrying out indirect desulfurization mind with the degree of vacuum of 20torr for 20 minutes, it moved to oven, deaerated, and the vacuum press was carried out, holding for 30 minutes at 90 more °C. Thus, sticking by pressure was performed for the glass laminate by which preparative pressure arrival was carried out for 20 minutes on condition of 135 °C and pressure 1.2MPa in autoclave, and the glass laminate was obtained.

[0048] It carried out like Example 1 except using the ITO distribution plasticizer which made comparative example 5 triethylene-glycol di-2-ethyl HEKISHI rate 20 weight section distribute ITO particle 5 weight section.

It carried out like Example 1 except using the sheet silicate distribution plasticizer which made comparative example 8 triethylene-glycol di-2-ethyl HEKISHI rate 20 weight section distributes sheet silicate 23 weight section.

The glass laminate was produced using the usual interlayer (average 0.76 mm in thickness) which does not contain comparative example 9ITO and a sheet silicate as heat reflective glass with which lamination coating of a silver film and the thin film of a metallic oxide was carried out in one sheet of the float glass used when producing a glass laminate.

By two usual interlayers (average 0.38 mm in thickness) which do not contain comparative example 10ITO and a sheet silicate, the silver film and the thin film of the metallic oxide put heat ray reflective PET by which lamination coating was carried out, inserted with the transparent float glass from both ends further, and created the glass laminate.

from area under the curve) and showed the glass laminates [0049][Evaluation]. The following evaluation was performed about the interlayer for glass laminates and glass laminate which were obtained by the above-mentioned example and the comparative example, and the result was summarized in Tables 2 and 3.

1) The transmissivity of 300–2500 nm of a glass laminate is measured using an optical property Naoki spectrophotometer (Shimadzu "UV3100"). It asked for the visible light transmittance ( $T_v$ ) of 380–780 nm, and the solar transmittance ( $T_s$ ) of 300–2500 nm by JIS Z 8722 and JIS R3106 (1988).

2) Haves of the glass laminate was measured based on Haves JIS K 6714.

[0050]3 The transmission electron microscope (TEM) was used after super-flake production of the dispersion state interlayer in the inside of the film of ITO particles, and the dispersion state of ITO particles was photoed and observed. The particle diameter of ITO particles was made into the longest path of the ITO particles in the photograph obtained by the above-mentioned photography. The particle diameter of all the ITO particles in 10 micrometers x 10 micrometers of the above-mentioned photographing areas was measured, and it asked for mean particle diameter with a volume conversion average. It asked for the particulate number with a particle diameter of not less than 100 nm which exists in the above-mentioned photographing area, and the number of per 1 micrometer<sup>2</sup> was counted.

(1) A viewing device, a condition and a transmission electron microscope:H-7100FA type (made by Hitachi Ltd.)

— accelerating voltage: 100-kV(2) section manufacturing device and ultramicrotome: EM-ULTRACUT-5 (made by the Leica company)

- Freezing cutting system: REICHERT-NISSEI-FCS (made by the Leica company)

- Knife: DIATOME ULTRA CRYO DRY (made by DIATOME)

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especially Hayes are good, and] for glass laminates which can adjust adhesive strength, and the glass laminate using the interlayer can be provided.

**[Translation done.]**

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